Evaluation of Cefuroxime Axetil Drug as Acid Corrosion Inhibitor for Zinc using Polarization and Weight Loss Technique

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Abstract: *The corrosion inhibition of zinc in 1.0 M H_2SO_4 solution by Cefuroxime axetil has been studied in relation to the concentration of the additive using weight loss measurement, Linear polarization resistant (LPR) and potentiodynamic polarization (PDP).The results indicate that Cefuroxime axetil inhibited corrosion reaction in the acid medium and inhibition efficiency increased with inhibitor concentration. The inhibitor was found to be adsorption inhibitor that best fitted the Langmuir adsorption model. From calculated values of activation energy, free energy of adsorption and the variation of inhibition efficiency with temperature, it was deduced that the adsorption of the inhibitor favours the mechanism of physical adsorption. The polarization data indicate that the inhibitor was of mixed type. The results obtained from LPR, PDP and weight loss measurements were in good agreement.*

Keywords: *Adsorption, Corrosion inhibition, polarization, Cefuroxime axetil.*

1. INTRODUCTION

The use of inhibitors is one of the most universal and economical measures to combat corrosion of metals [1 – 2]. When present in small quantities in an aggressive medium they retard corrosion by bringing about changes in the surface condition of a metal.Organic inhibitors have been found to have wide applications in cleaning solutions for industrial equipment, as components in pre-treatment compositions and in acidisation of oil wells owing to their less toxicity and eco friendliness [3 – 4]. Most organic inhibitors are facilitated by the presence of polar groups in the molecular structure which can strongly adsorb on to the metal surface or react with the corrosion product on the surface to form a protective layer [5]. The most effective polar groups include sulphur, nitrogen, oxygen and phosphorous atoms.

Drugs come in a wide variety of molecular weights with carboxylic and / or heterocyclic systems making many of them suitable for use as corrosion inhibitors [6 - 9]. The corrosion protection properties of the b-lactam group (largest group of antibacterial agents used in clinical medicine) of antibiotics have attracted too much attention in recent years [10]. Structurally, the group have a basic unit of a four-membered nitrogen-containing b-lactam ring that gives these agents their antibacterial activity and makes it suitable to be used as inhibitors. They are divided into four groups on the basis of the molecular structures surrounding and supporting this active site viz: penicillins, cephalosporins, carbapenems and mono bactams,. Of this group, there is scanty literature proving the corrosion inhibition efficiency of second generation cephalosporins (cefaclor, cefoxitin, cefprozil, and cefuroxime).

The second generation cephalosporins have a greater gram-negative spectrum while retaining some activity against gram-positive bacteria. They are useful agents for treating upper and lower respiratory tract infections, sinusitis and otitis media. These agents are also active against E. coli, Klebsiella and Proteus, which makes them potential alternatives for treating urinary tract infections caused by these organisms [11].

We have reported recently the adsorption characteristics and corrosion inhibition ability of Cefuroxime Axetil a second generation cephalosporins for Aluminum in Hydrochloric Acid Solution using thermometric, gasometric weight loss and scanning electron microscope (SEM) techniques [9]. The inhibition of aluminium corrosion by Cefuroxime Axetil was attributed to the presence of heteroatoms (nitrogen oxygen and sulphur atom) as reactive center through which it adsorb on the metal surface. The encouraging results obtained by this research permit to us to further test the activity of the drug in another media (sulphuric acid).

The present study presents the appraisal of inhibitive capability of Cefuroxime axetil on zinc in 0.1 M H_2SO_4 by using weight loss measurement, linear polarization resistant (LPR), and potentiodynamic polarization (PDP).

2. METHODOLOGY

2.1. Gravimetric (Weight Loss) Method

Gravimetric methods were carried out at 303, 313, 323 and 333 K for 7 days using the method described in literature [12]. From the average weight loss (mean of three replicate analyses) results, the inhibition efficiency (% I) of the inhibitor, the degree of surface coverage (θ) and the corrosion rate of Zinc (CR) were calculated using equations 1 to 3 respectively [13],

$$\%I = \frac{w_1 - w_2}{w_1} \times 100 \qquad 1$$

$$\theta = \left[1 - \frac{w_1}{w_2}\right] \qquad 2$$

$$CR = \frac{w_1 - w_2}{At} \qquad 3$$

where w_1 and w_2 are the weight losses of Zinc (in grams) in the absence and presence of the inhibitor respectively. θ is the degree of surface coverage of the inhibitor, CR is the corrosion rate of zinc, A is the surface area of the Zinc coupon in cm^2 and t is the period of immersion in hours.

2.2. Potentiodynamic Polarisation

The potentiodynamic current-potential curves were recorded by changing the electrode potential (E_{corr}) automatically with a scan rate 0.33 mV s^{-1} from a low potential of -800 to -300 mV (SCE). Before each run, the working electrode was immersed in the test solution for 30 min to reach steady state.

The corrosion rate of the structure shall be calculated through corrosion current density (l_{corr}). The linear Tafel segments of the anodic and cathodic curves obtained were extrapolated to corrosion potential to obtain the corrosion current densities (i_{corr}). The inhibition efficiency (1 %) was evaluated from the measured i_{corr} values [14].

$$\%I = \frac{i^0_{Corr} - i_{Corr}}{i^0_{Corr}} \times \frac{100}{1} \qquad 4$$

where i^0_{Corr} and i_{Corr} are the uninhibited and inhibited corrosion current densities, respectively.

2.3. Linear Polarisation Resistance

Linear Polarisation Resistance measurements were be carried out within the potential range ± 20mV with respect to the open circuit potential, and the current response was measured at a scan rate of 0.5 mV/S. The over potential and current data was plotted on a linear scale to get linear polarisation resistance (LPR) plots, and the slope of the plots in the vicinity of the corrosion potential gave the polarisation resistance (R_p).

From the measured R_p values, the inhibition efficiency (I %) was calculated using equation 7 [14]:

$$\%I = \frac{R_{p(Inh)} - R_p}{R_{p(Inh)}} \times \frac{100}{1} \qquad 5$$

where R_p and $R_{p(inh)}$ are the uninhibited and inhibited polarization resistance, respectively

3. RESULTS AND DISCUSSION

3.1. Gravimetric Study

Plots obtained from weight loss measurement of zinc in 0.1 M H_2SO_4 in the presence and absence of Cefuroxime axetil at various concentrations after immersion for 168 hours at 303K is shown in Figure 1.From the plot, it is evident that weightloss of zinc in solutions of H_2SO_4 increases with
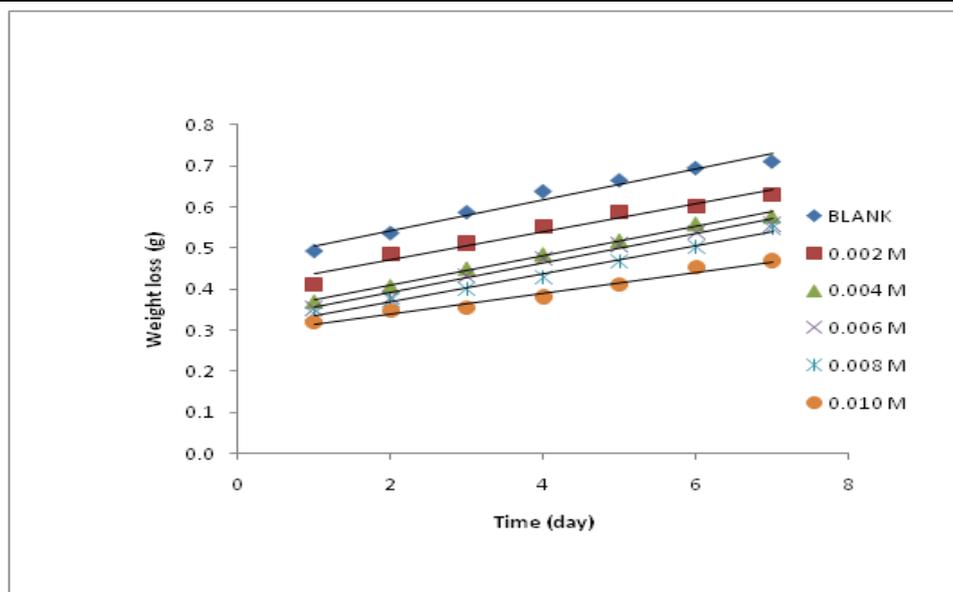
Figure1. *Variation of weight loss with time for the corrosion of zinc in 0.1 M H_2SO_4 containing various concentrations of Cefuroxime Axetil at 303K*

Time but decreases with increasing concentration of the inhibitor indicating that the rate of corrosion of zinc in solutions of H_2SO_4 is retarded by the presence of inhibitor [15 – 16]. At higher temperature (plots not shown), it was indicative that weight loss of zinc increases with increasing temperature hence the drug is adsorbed on zinc surface through the mechanism of charge transfer from charged inhibitor to charged metal surface, which supports physical adsorption.

Values of inhibition efficiencies and corrosion rate of Cefuroxime axetil calculated using equation 1 and 3 respectively is presented in Table 1.

Table1. *Corrosion rate of zinc in solution of 0.1 M H_2SO_4 and inhibition efficiencies of Cefuroxime axetil for zinc in 0.1 M H_2SO_4*

System (M)		Inhibition efficiency (%)			Corrosion rate ($gh^{-1}cm^{-2}$)			
	333 K	323 K	313 K	303 K	333 K	323 K	313 K	303 K
Blank	-	-	-	-	0.00850	0.00703	0.00657	0.00591
0.002	34.25	40.76	47.70	56.02	0.00341	0.00304	0.00278	0.00221
0.004	39.82	42.90	52.18	59.97	0.00311	0.00295	0.00249	0.00202
0.006	41.32	45.99	54.53	64.25	0.00280	0.00276	0.00234	0.00183
0.008	45.19	47.70	54.21	66.70	0.00269	0.00267	0.00235	0.00174
0.01	49.55	51.65	59.44	69.37	0.00258	0.00248	0.00215	0.00155

The results obtained generally revealed that the inhibition efficiencies of the inhibitor increases with increase in concentration, indicating that Cefuroxime axetil is an adsorption inhibitor. The inhibition efficiencies were also found to decrease with increasing temperature, which implies that the extent of adsorption also decreases with increase in temperature. Hence the inhibitor acted through the mechanism of physical adsorption, which involves charge transfer from charged inhibitor to charged metal surface [17].

It is also evident from the values of corrosion rates calculated and presented in Table 2
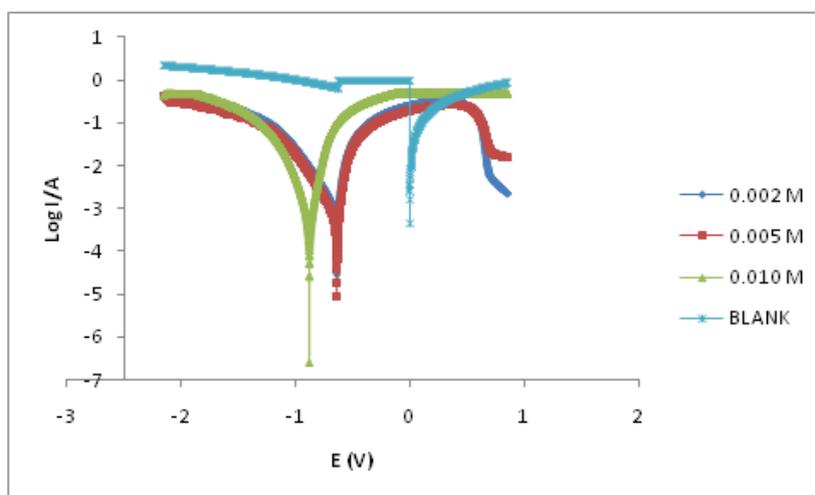
Table2. *Potentiodynamic polarization resistant data for the corrosion of zinc in 0.1 M H$_2$SO$_4$ containing various concentrations of Cefuroxime axetil at 303 K*

Conc. of inhibitor (M)	E$_{corr}$ (mV)	I$_{corr}$ (µA)	β$_a$ mVdec^{-1}	β$_c$ mVdec^{-1}	θ	IE %
Blank	- 563	353. 50	163.0	172.5		
0.02	-1144	100.24	142.5	153.5	0.7164	71.64
0.05	-894	78.26	135.1	146.1	0.7853	78.53
0.1	-659	60.51	130.0	136.5	0.8288	82.88

that as the temperature of the system is increased, the corrosion rates of zinc increase at all the concentrations of Cefuroxime axetil. The corrosion rate also decreased with increase in the concentration of the inhibitor.This observation implies that the surface area of the zinc covered by the inhibitor increases as the concentration of the inhibitor increases.

3.2. Polarization Study

Linear polarization resistant (LPR) and potentiodynamic polarization (PDP) studies were also conducted during the study. From PDP study, plots obtained for the variations of applied potential with log (current) for corrosion of zinc in the absence and presence of inhibitor is depicted in Figure2.

Figure2: *Potentiodynamic polarization curves for the zinc steel in 0.1M H$_2$SO$_4$ in the absence and presence of different concentrations of Cefuroxime axetil*

It can be observed from the plots that addition of inhibitor inhibits corrosion as indicated by the systematic displacement of the curves towards lower current densities with increasing inhibitor concentration. Electrochemical parameters such as corrosion current density (I$_{corr}$), corrosion potential (E$_{corr}$), anodic and cathodic Tafel slopes (β$_a$ and β$_c$), the degree of surface coverage (θ) and the percentage of inhibition efficiency (IE %) deduced from the plots are presented in Table 2.

Table3: *Linear polarization resistant (LPR) parameters for the corrosion of zinc in 0.1 M H$_2$SO$_4$ containing various concentrations of Cefuroxime axetil at 303 K*

C (M)	R$_p$ (Ω/cm^2)	% I
Blank	70.55	-
0.02	168.05	58.01
0.04	227.43	68.98
0.06	275.05	74.35
0.06	305.45	76.90
0.1	393.38	82.07

presents values of polarization resistant as well as calculated inhibition efficiencies obtained from linear polarization study. The values also show appreciable correlation with data obtained from weight loss and PDP measurements.

3.3. Kinetic study

The kinetic of the corrosion of zinc in H$_2$SO$_4$ containing various concentrations of Cefuroxime axetil was studied by fitting weight loss data to different kinetic models. The test revealed that by plotting – log (weight loss) versus time, linear plots (Figure 3)

Evaluation of Cefuroxime axetil drug as acid corrosion inhibitor for zinc using polarization and weight loss technique
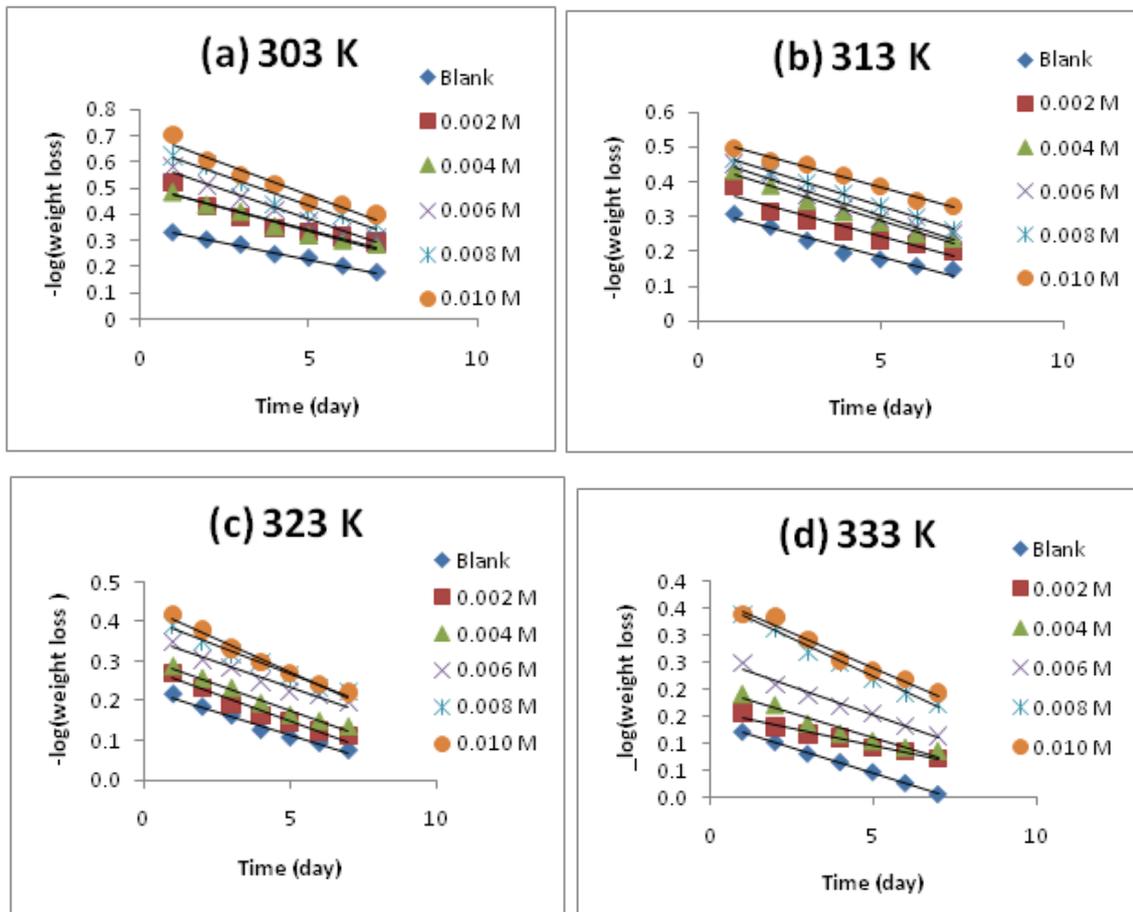

Figure3. *Variation of –log(weight loss) with time for the corrosion of zinc in 0.1 M H₂SO₄ containing various concentrations of Cefuroxime axetil at 303, 313, 323 and 333 K*

Table4. *Kinetic parameters for the corrosion of in 0.1M H₂SO₄ containing various concentrations of Cefuroxime axetil*

Temperature	System (M)	slope	k_1	$t_{1/2}$	R^2
303 K	Blank	0.0485	0.111696	6	0.9944
	0.002	0.0456	0.105017	7	0.8938
	0.004	0.0449	0.103405	7	0.9712
	0.006	0.0345	0.079454	9	0.9787
	0.008	0.0345	0.079454	9	0.9535
	0.01	0.0254	0.058496	12	0.9518
313 K	Blank	0.0340	0.08955	8	0.9495
	0.002	0.0353	0.086221	8	0.9496
	0.004	0.0331	0.076229	9	0.9827
	0.006	0.0346	0.079684	9	0.9705
	0.008	0.0336	0.077381	9	0.9985
	0.01	0.0285	0.065636	11	0.9831
323 K	Blank	0.0333	0.07669	9	0.9833
	0.002	0.0265	0.064945	11	0.9632
	0.004	0.0268	0.06103	11	0.9803
	0.006	0.0249	0.06172	11	0.9607
	0.008	0.0282	0.057345	12	0.9849
	0.01	0.0333	0.056342	12	0.9825
333 K	Blank	0.0283	0.065175	11	0.9170
	0.002	0.0259	0.059648	12	0.9687
	0.004	0.0232	0.049954	14	0.9503
	0.006	0.0212	0.048824	14	0.9809
	0.008	0.0200	0.046556	15	0.9892
	0.01	0.01980	0.045153	15	0.9749

were obtained indicating that the corrosion of Zinc in H_2SO_4 is first order and that equation 8 is applicable [13].

$$-log \, (weight \, loss) \, = \, k1t/2.303 \qquad\qquad 8$$

where k_1 is the rate constant for the first order reaction and t is time (in days).

Values of R^2, slopes and k_1 deduced from the plots are presented in Table 4. The half life of a first order reaction ($t_{1/2}$) is related to the rate constant according to the following equation [19],

$$= \frac{0.6930}{k_1} \qquad\qquad 9$$

From the results obtained (Table 4), It is evident that the half life for the inhibited reactions of Zincin solutions of H_2SO_4 are higher than those obtained for the blank indicating that Cefuroxime axetil has the tendency to increase the half-life of zinc in solutions of H_2SO_4.

3.4. Effect of Temperature

In acidic solution, the corrosion rate is related to temperature by modified Arrhenius equation [20]

$$logCR \, = \, logA - \frac{E_a}{2.303R}\left(\frac{1}{T}\right) \qquad\qquad 10$$

Where CR is the corrosion rate determined from the weight loss measurement, Ea, the apparent activation energy, A the Arrhenius constant, R the molar gas constant and T is the absolute temperature. The apparent activation energy was determined from the slope of log CR versus I/T graph depicted in Figure 4
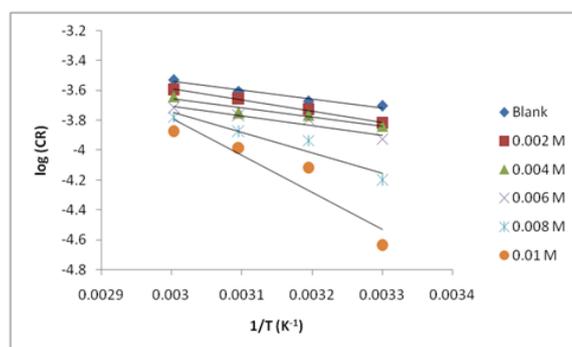
Figure4. *Arrhenius plot for the corrosion of zinc in 0.1 M H_2SO_4 containing various concentrations of Cefuroxime axetil*

$$log\frac{C}{\theta} \, = \, logC - logb \qquad\qquad 11$$

The values of activation energies were calculated and given in Table 5. These values indicate that the presence of studied inhibitor increases the activation energy of the metal dissolution reaction. The adsorption of the studied inhibitors is assumed to occur on the higher energy sites and the presence of the inhibitor, which results in the blocking of the active sites, must be associated with an increase in the activation energy of zinc corrosion in the inhibited state [21]. The higher value of E_a in the presence of inhibitor compared to that in its absence is interpreted as an indication of Physisorption [13]

$$. \, logb \, = \, -1.744 \, - \frac{\Delta G_{ads}}{2.303RT} \qquad\qquad 12$$

Table5. *Arrhenius and transition state adsorption parameters for the corrosion of zinc in 0.1 M H_2SO_4 containing various concentration of Cefuroxime axetil*

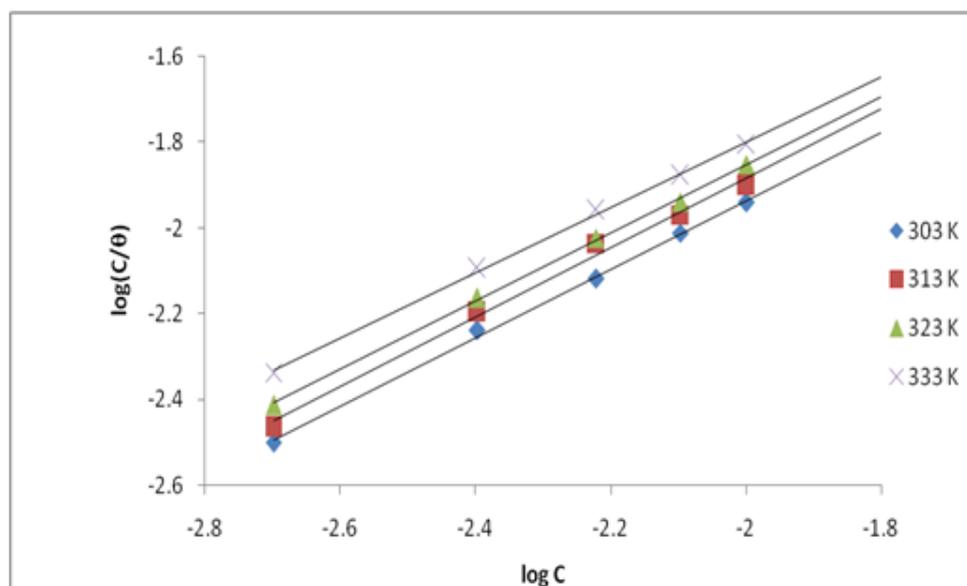| C(M)	Arrhenius parameters			Transition state parameters				
	log A	E_a (J/mol)	R^2	slope	intercept	ΔS^0_{ads} (J/mol)	ΔH^0_{ads} (J/mol)	R^2
Blank	-744.0	5.88	0.960	1.034	-10.85	976.60	-8.59667	0.936
0.002	-653.2	26.02	0.994	1.094	-9.89	1001.49	-11.5980	0.991
0.004	-613.2	33.41	0.918	1.186	-9.72	998.77	-9.09551	0.872
0.006	-587.1	34.06	0.912	1.395	- 9.69	1043.07	-9.86040	0.864
0.008	-246.4	34.79	0.912	1.579	-8.89	1047.92	-13.1278	0.893
0.010	-135.1	59.02	0.876	1.735	-8.75	1099.49	-14.4247	0.862

3.5. Thermodynamic Consideration

The values of enthalpy of activation ΔH_{ads}^{o} and entropy of activation ΔS_{ads}^{o} were obtained from the transition state equation [20]

$$CR \ = \ \frac{RT}{Nh} exp\left(\frac{\Delta S_{ads}^{0}}{R}\right) exp\left(\frac{-\Delta H_{ads}^{0}}{RT}\right) \qquad\qquad 13$$

where N is the Avogadro's number, h is the plank constant, ΔS_{ads}^{0} is the standard entropy change, ΔH_{ads}^{0} is the standard enthalpy change, R is the gas constant and T is the temperature. Rearrangement of equation 13 and taking the logarithm of both sides of the equation, yielded equation 14.

$$ln\left(\frac{CR}{T}\right) \ = \ ln\left(\frac{R}{Nh}\right) \ + \ \frac{\Delta S_{ads}^{0}}{R}$$
$$- \frac{\Delta H_{ads}^{0}}{RT} \qquad\qquad 14$$

A plot of ln (CR/T) as a function of 1/T (Figure 5)

Figure5. *Langmuir Isotherm for the Adsorption of Cefuroxime Axetil on Zinc Surface*

was made and straight line was obtained. ΔH_{ads}^{o} and ΔS_{ads}^{o} was computed from the slope and intercept respectively, from the linear plot. The computed values of the thermodynamics parameters of activation for the dissolution of zinc at different temperatures are presented in Table 5.

Examination of this data reveals that the values of ΔH_{ads}^{o} and ΔS_{ads}^{o} in the presence of Cefuroxime axetil increase over that of the uninhibited solution. This implies that energy barrier of the corrosion reaction in the presence of inhibitor increases which is expected. The negative values of ΔH_{ads}^{o} reflect the exothermic behaviour of the adsorption of studied inhibitor on the zinc surface. The ΔS_{ads}^{o} value is positive, which are opposite to the usual expectation that the adsorption is an exothermic process and always accompanied by a decrease of entropy. The reason can be explained as follows: the adsorption of organic inhibitor molecules from the aqueous solution can be regarded as a quasi-substitution process between the organic compound in the aqueous phase [org (sol)] and water molecules at the electrode surface [$H_2O_{(ads)}$] [21]. In this situation, the adsorption of studied inhibitor is accompanied by desorption of water molecules from the electrode surface. Thus, while the adsorption process for the inhibitor is believed to be exothermic and associated with a decrease in entropy of the solute, the opposite is true for the solvent. The thermodynamic values obtained are the algebraic sum of the adsorption of organic inhibitor molecules and desorption of water molecules. Therefore, the gain in entropy is attributed to the increase in solvent entropy [20]. The positive values of ΔS_{ads}^{o} suggest that

the adsorption process is accompanied by an increase in entropy, which is the driving force for the adsorption of studied inhibitor on the Zinc surface.

3.6. Adsorption Consideration

The adsorption process is influenced by the nature and surface charge of the metal, by the type of aggressive electrolyte and by the chemical structure of the inhibitors [22]. The gravimetric data from the present study were fitted to the most frequently used adsorption isotherms, namely: Temkin, Frumkin, Langmuir, Freundlich, Hill de Boer, Parsons, Flory Huggins, Dhar-Flory Huggins, and Bockris Swinkles, and the correlation coefficients (R^2) were used to determine the best fits. The experimental data fit best for the Langmuir adsorption isotherm as the correlation coefficients (R^2) were on the range $0.999 \geq R^2 \geq 0.992$. This suggests that the adsorption of studied inhibitors on the metal surface obeyed the Langmuir adsorption isotherm which is given as equation 11 [23].

Where is the concentration of the inhibitor in the bulk electrolyte,

θ is the degree of surface coverage of the inhibitor and b is the equilibrium constant of adsorption. Figure 6
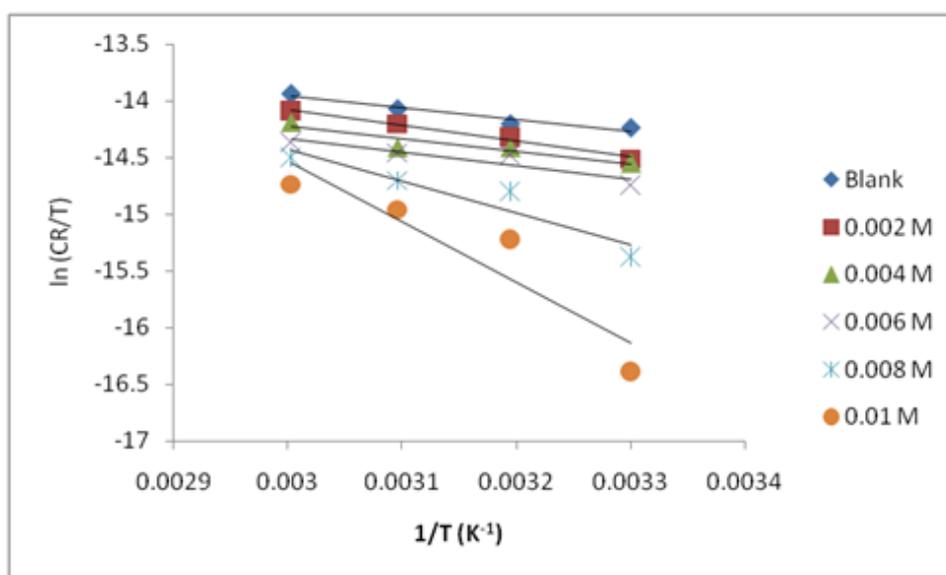
 Figure6. *Transition state plot for the corrosion of zinc in 0.1 M H$_2$SO$_4$ containing various concentrations of Cefuroxime Axetil*

shows the Langmuir isotherm for the adsorption of Cefuroxime axetil on zinc surface. Adsorption parameters deduced from the isotherm are presented in Table 6.

Table6: *Langmuir parameters for the adsorption of Cefuroxime axetil on the surface of zinc*

T (K)	slope	log b	ΔG^0_{ads} ($\frac{kJ}{mol}$)	R^2
303	0.9244	0.798	-14.75	0.998
313	0.8834	0.809	-15.30	0.992
323	0.8766	0.793	-15.69	0.999
333	0.8432	0.761	-15.97	0.998

It is a known fact that b$_{ads}$ denotes the strength between adsorbate and adsorbant [24]. Large values of b$_{ads}$ imply more efficient adsorption and hence better inhibition efficiency [25]. It is clear from the table that values of b$_{ads}$ are very low indicating weak interaction between the inhibitor and the zinc surface. It seems therefore, that electrostatic interaction (Physisorption) between the inhibitor molecules existing as cations should prevail over molecular interaction which often results in strong interactions (Chemisorption) [26].

The equilibrium constant of adsorption obtained from the Langmuir adsorption isotherms is related to the standard free energy of adsorption according to the following equation [9], where ΔG^0_{ads} is the standard free energy of adsorption,R is the gas constant, b is the equilibrium constant of adsorption and T is the temperature

Values of free energy calculated from equation 12 are presented in Table 6. In general, the negative values of the ΔG_{ads} reflect that the adsorption of the inhibitors on zinc surface from the 0.1 M H_2SO_4 is spontaneous process [20]. The free energies (which ranged from -15.97 to -14.75 kJ/mol) are negatively less than the limit (-40 kJ / mol) expected for the mechanism of chemical adsorption further affirming that the adsorption of Cefuroxime axetil on Zinc surface is spontaneous and supports physiosorption.

4. CONCLUSION

From the results and findings of this study, it can be concluded that Cefuroxime axetil is a good adsorption inhibitor for Zinc in acidic medium. The inhibitor acted through the mechanism of physical adsorption and their adsorption behaviour is best described by the Langmuir adsorption models. Data obtained from electrochemical and gravimetric are in good agreement with each other.

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